RAPID AND EFFECTIVE METHODS FOR THE SCREENING OF FLAX FIBRES FOR COMPOSITE APPLICATIONS

K. Hendrickx[1], R. D. Romian[1], T. Goedemé[2], A.W. Van Vuure[1], J. Ivens[1]

[1]Department of Materials Engineering, KU Leuven
Kasteelpark Arenberg 44, box 2450, B-3001 Leuven, Belgium
Email: kevin.hendrickx@kuleuven.be, web page: http://www.mtm.kuleuven.be

[2]Department of Electrical Engineering, KU Leuven
Kasteelpark Arenberg 10, box 2441, B-3001 Leuven, Belgium
Email: toon.goedeme@kuleuven.be, web page: http://www.esat.kuleuven.be

Keywords: Flax fibres, fineness, purity, screening

ABSTRACT

The effect of technical flax fibre fineness, purity and strength on the composite performance was investigated in order to provide screening methods to assess the suitability of the fibres for composite applications. Fineness significantly affects the transverse flexural strength of unidirectional composites while the effect of fineness and purity on the tensile performance is limited. It was also found that properties extracted from single fibre tensile tests on flax fibres do not correlated with back-calculated properties from composite tests. It is therefore advised to use composite testing rather than relying on tests on single technical fibres or fibre bundles to predict the composite performance of the fibres.

1 INTRODUCTION

The use of flax fibres in composites has steadily increased over the last decade. However, screening methods for flax fibres are still mainly oriented towards textile applications. It is reasonable to assume that fibre requirements differ significantly depending on the application field. Consequently, rapid screening methodologies are needed to assess the suitability of these fibres for composite applications.

Flax fibre composite properties are most likely affected by three main fibre characteristics which are intrinsically coupled: fineness, purity and strength [1-3]. Finer fibres have a larger surface in contact with the resin for a specific fibre volume. This is expected to increase composite performance, taking into account the weak bundle cohesion of technical flax fibres [4]. Increased fineness is often achieved by intensive hackling and/or carding operations offering the additional advantage of removing impurities attached to the fibre bundles. The main impurities in flax fibre reinforcements are the woody particles, designated as shives, which are remnants from the flax plant tissues that did not detach from the bundles during scutching and hackling. It has been established that flax shive inclusion leads to a decrease in mechanical properties of the resulting composites [5]. From the previous it could be expected that very fine and pure fibres would ensure optimal composite performance. Unfortunately by increasing fineness and purity, the refining will induce damage in the form of kink bands to the fibres [6].

To present it is unclear how the above affects composite performance. This work tries to clarify the relationships between fibre properties and their composite behaviour. Moreover, simple screening methods are proposed to evaluate the fibre for composite application purposes.

2 MATERIALS AND METHODS

2.1 Fibres and composite production

Scutched flax fibres were provided by three flax producing companies: Wavalin, Vanacker Rumbeke and Verhalle. The retting and scutching parameters are known for each batch. Under retted (UR), dew retted (DR) and over retted (OR) fibres were examined on fineness, purity and strength. Hackled fibres were acquired from Lineo NV as FlaxTape 200, a unidirectional flax tape with an areal density of 200 g/m².

A thermosetting bisphenol A resin, Epikote 828 LVEL, was used to impregnate the flax fibres. To initiate the crosslinking reaction, 15.2 g of 1,2-diamino cyclohexane (Dytek DCH-99) was added per 100 g of resin.

The pure matrix material has a strength of 75 MPa, a stiffness of 2.87 GPa and a strain to failure of 3.9% [7].

All composite samples were produced by vacuum assisted resin infusion (VARI). Flax fibres were dried for 24 h at 60°C. Unidirectional tensile test specimen dimensions were 1 cm x 20 cm x 2 mm and were produced to size. As-received scutched fibres were incorporated in the resin without additional alignment or combing steps. The desired fibre volume fraction was set to 40% and was controlled by weighing the flax fibres prior to impregnation by the resin. Three point bending samples were machined from 2.5 cm x 20 cm x 2 mm unidirectional composites with a wet diamond saw. The final dimensions of the samples was 2.5 cm x 1 cm x 2 mm.

2.2 Fineness evaluation

Three methods were used to determine the fineness of the fibres: the commonly used and standardized Airflow method, a gravimetric method, and an optical method.

Airflow measurements were carried out by Celabor s.c.r.l on samples weighing 2.5 g in accordance with the ISO 2370 standard. The airflow rate was constant and equal to $20x10^{-3}$ m³/min. The device was calibrated using standard flax fibre samples with IFS values ranging from 21.7 to 72.1.

Gravimetric analysis yields the mass of the fibre per unit length, from which an average fibre cross section can be calculated. When extracting a technical fibre from the batch, care was taken to preserve its structural integrity. Technical fibres were weighed using an analytical balance with an accuracy of 0.01 mg. Assuming a uniform cross-section of the technical fiber along its length, average cross-sections were determined; if in addition a circular geometry of the cross-section is assumed, the equivalent technical fiber diameters can be calculated. The density of the fibres was assumed to be 1.44 g/cm³.

For the optical characterisation method, an image processing algorithm was developed that analyses the distribution of the fibre thickness in a sample. The algorithm is fed with images of the fibre sample to be examined, taken with a commodity flatbed scanner with a resolution of 1200 dpi. The following image processing steps are executed in order to analyse the sample. Firstly, the image is cropped out and thresholded with a user adjustable threshold, yielding a binary image of the fibres. Then, this image is cleaned up by a sequence of morphological opening and closing operations, with a circular kernel with a user defined diameter. Subsequently, the Euclidean distance transform is executed on this cleaned-up binary image, in which for each fibre pixel the distance to the nearest background pixel is computed. Thereafter, local maxima of the distance transform output are computed, which are the pixel locations of the centres of the fibres. The actual value of the distance transform output at each of these positions is treated as a sample measurement of the (half) fibre thickness. The statistical distribution of the fibre thicknesses in the image is then computed by a histogram operation of these collected thickness values. Actual values of the pixel thickness can be computed out of these pixel numbers by taking into account the resolution of the scanned image.

2.3 Purity evaluation

Flax fibre purity was assessed gravimetrically using an analytical balance, accurate to 0.01 mg. Scutched fibres were manually combed in three stages using a different comb in each stage. For each stage 100 combing operations were performed. The combs differed in pin spacing, which decreased from 18 mm to 5.8 mm and finally to 2.2 mm. The latter two combs had two equally spaced but staggered rows of pins while the former had only one row of arch-like protrusions. During combing, material that became separated from the fibres, including short fibres, dust and shives, was collected and considered as impurity. The fibres after the third combing step were assumed to be free from impurities. The degree of purity was then defined as the ratio of clean fibre mass to fibre mass after a certain combing step.

2.4 Mechanical testing

2.4.1 Fibre

Single technical fibres were manually extracted from the scutched fibre bundles. Fibres showing signs of fibrillation, i.e. splitting into several smaller technical fibres, were excluded from further measurements. The equivalent diameter of each individual fibre was determined gravimetrically with an analytical balance, accurate to 0.01 mg. C-shaped frames were cut from sand paper with a grain size of 1000 so that the gauge length of the fibres was 50 mm. Technical flax fibres were glued to the frames, at both ends, with a thin, low strength adhesive to ensure the sand paper grains were responsible for fibre gripping. White markers, approximately 3 mm in diameter, were attached to the fibres in the gauge length region. These markers were sprayed with black paint to produce a speckle pattern, suitable for optical strain registration. Tensile tests were carried out on an Instron 5985, equipped with a 100 N load cell. Strain was registered optically with a digital image correlation set-up of Limess GmbH and results were processed with Vic2D software.

2.4.2 Composite

An Instron 4467, equipped with a 30 kN load cell, was used to test the composite samples in tension. Strain was registered with a 50 mm gauge length extensometer. Sand paper was used to prevent slippage of the sample in the clamps. The crosshead displacement rate was set to 1 mm/min.

Three point bending tests were carried out on an Instron 4467, equipped with a 1 kN load cell. The crosshead displacement rate was 1 mm/min. The span to thickness ratio was only 8:1 due to difficulties in achieving fibre alignment of the scutched material over large span lengths. However, the transverse tensile strength is expected to be significantly lower than the transverse shear strength, thus the effect of the shear stresses can be neglected [4].

Single fibre tests were executed at a relative humidity level of 50% and a constant temperature of 20°C. Composite samples were conditioned for at least 7 days at these conditions prior to testing.

3 RESULTS AND DISCUSSION

3.1 Fineness evaluation

Producers of flax fibres often make predictions of fibre fineness after further processing based on a qualitative estimate of the degree of retting of the fibres. However, dividing the fibres into three types of retting categories, namely under retted, dew retted and over retted, based on their appearance and scent makes this method extremely subjective, as will be shown later.

Airflow measurements were carried out as a first screening method to determine the fibre fineness. A major disadvantage of this technique is that no information on the fineness distribution is produced during the measurements but merely an average fineness value is obtained. The fineness distribution could also have an important impact on composite performance. The results in figure 1 show that the retting categorization of flax fibres does not correlate with IFS. It was expected that over retted fibres would lead to the smallest IFS values as bundle disintegration is facilitated by the microbial degradation of polysaccharides which are responsible for the bundle cohesion. As shown in figure 2 the correlation coefficient between gravimetric measurements and airflow values was only 0.51. This implies that IFS values cannot be directly related to fibre diameter, which is again a major shortcoming of this technique.

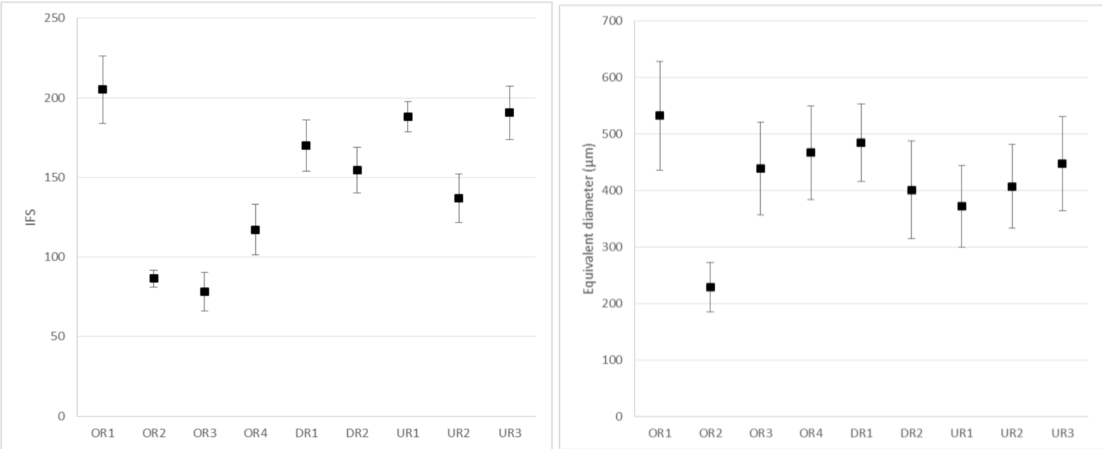


Figure 1: Left: Airflow IFS values show that categorizing fibres according to retting degree is subjective. Right: Gravimetrical analysis is not ideal to determine flax fibre fineness because sampling introduces large uncertainties. All error bars represent standard deviations.

Technical fibre sampling is the most determining factor in the gravimetrical analysis method. A technical fiber is by definition a conglomerate of elementary fibres. On one location the technical fiber can consist of a large amount of elementary fibers, but it can split up into multiple thinner technical fibers in another location along its length. A standardized method to identify and extract a technical fibre from the batch is therefore necessary but wasn't developed in the framework of this work leaving the technique operator dependent.

It was also seen that the correlation between IFS values from airflow measurements and the gravimetric equivalent diameters is low. This means that the sampling method in the gravimetrical method was suboptimal and/or the IFS is not directly linked to physical diameters but can also be influenced by other parameters such as fibre organization or the fineness distribution itself.

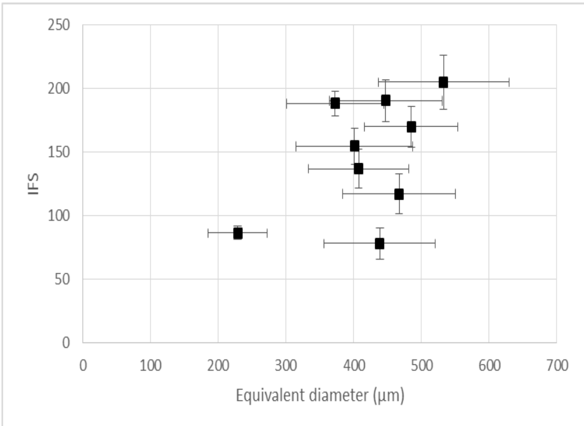


Figure 2: The correlation coefficient between IFS values and grametric equivalent diameters is only 0.51.

In view of the difficulties and uncertainties described above, it was decided to determine fineness by an automated image analysis algorithm which determines Ferret's diameter, i.e. the projected fibre diameter. This is an interesting technique since it enables the generation of a statistically reliable diameter distribution in a short time span. A proof-of-concept of this technique will be discussed in section 3.3.2.

3.2 Fibre properties

The effect of processing history on the fibre tensile properties will be evaluated in this section. Evidently, the effect of impurities will be very limited since they bear no load during tensile testing. The former assumption is only valid when the mass of the impurities is negligeable compared to the fibre mass which was verified by visual inspection. By extracting technical fibres that showed no sign of fibrillation from the batches, problems of uniform fibre loading during testing were avoided. As a result, fibre diameter variation was small, making it difficult to assess its influence on the tensile properties.

Single fibres from four different batches were selected for testing. The fineness of the fibre bundles, according to gravimetric analysis, was used to select these batches as an indirect representation of the scutching intensity. OR1 (VH-OR-14F) and OR3 (VA-OR-13B) were the coarsest and finenest batch respectively, whereas DR1 (VA-DR-13B) and UR1 (WRL-VR-12F) were intermediate batches. Figure 3 shows the results of the single fibre tests. Note that the stiffness is calculated in the region between 0.6% and 0.8% strain where the tensile modulus stabilizes as a function of strain.

From the data it is immediately clear that the variation on both stiffness and strength values within the different batches is large. From the data it appears that fibre stiffness is rather underestimated when compared to the values in composite materials, as will be shown later. As a general trend we can state that scutched fibres appear to possess a higher stiffness than when hackled fibres. This may be related to the presence of defects. However, results are contradictory since the batch with the largest fibre diameters, for which scutching was presumably less severe in terms of defects has a stiffness similar to the hackled material which is presumed to contain more defects. Only the dew retted fibre is signicantly stronger than the other fibres.

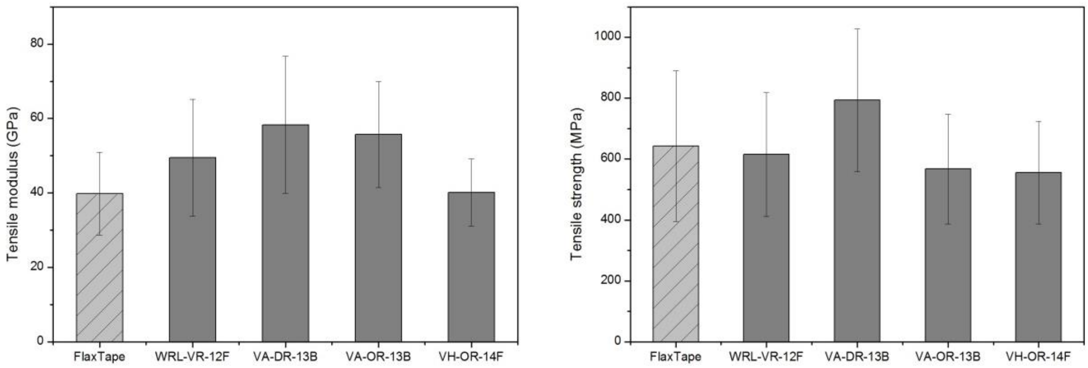


Figure 3: Left: Technical fibre tensile modulus, calculated between 0.6% and 0.8% strain. Right: Technical fibre tensile strength.

Figure 4 shows the evolution of tensile stiffness as a function of strain during fibre testing. For all materials a highly non-linear behaviour is observed, comparable to that of elementary fibres. When elementary flax fibres are loaded in tension their behavior is non-linear [8-10]. Processing steps such as scutching or hackling do not appear to have a significant influence on this behaviour, nor on the average strength and stiffness values. However, on the technical fibre level, the processing intensity of the fibres, i.e. the extent of mechanical loading during the processing, seems to play a major role.

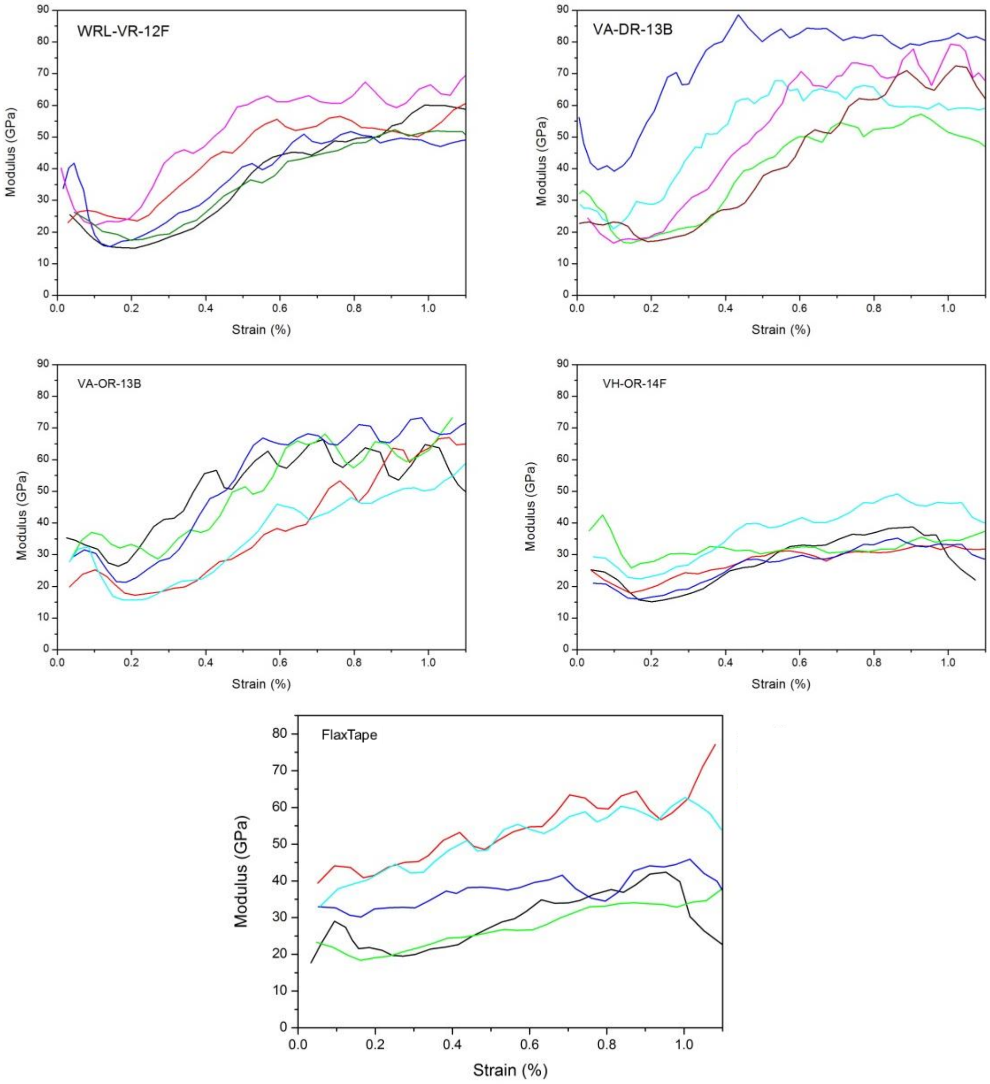


Figure 4: Evolution of technical fibre tensile stiffness as a function of strain for five different samples, illustrating the non-linear behavior of the fibres

Note that hackled fibres have a markedly different behaviour during tensile loading compared to scutched fibres. The investigation of the reasons for this difference are beyond the scope of this study but it is expected that it arises from the different defect densities present in the elementary and technical fibres of hackled and scutched flax fibres.

3.3 Composite properties

3.3.1 Longitudinal tension

Figure 5 shows the results of the tensile tests on the composites, normalized to a fibre volume fraction of 60%. Again, the stiffness was calculated between 0.6% and 0.8% strain enabling comparison with the results from single fibre tests. There was no significant difference found between the composite stiffness of the different batches. This is rather surprising considering the stiffness variation between the batches in figure 2.

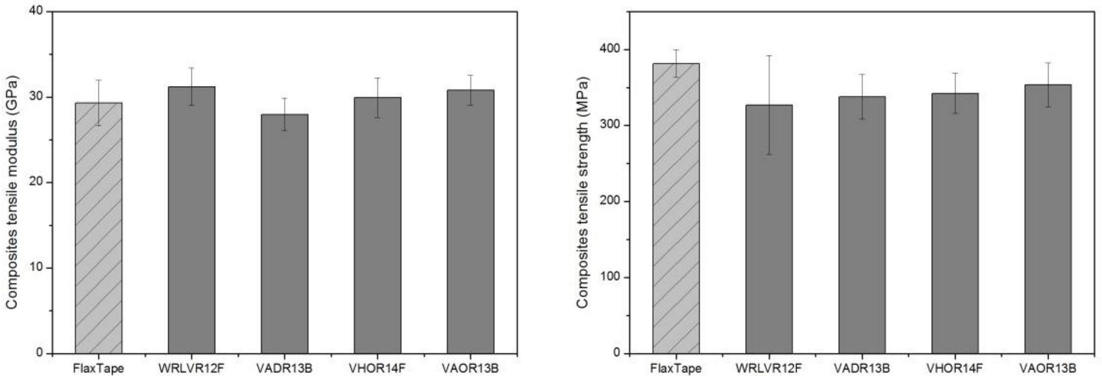


Figure 5: Left: Tensile modulus of composite samples, calculated between 0.6% and 0.8% strain. Right: Composite tensile strength.

Figure 6 shows the back-calculated fibre properties from the composite tests. It was expected that these values should correspond to the data shown in figure 2. However, comparing the data reveals large differences up to 14 GPa in absolute value. This result leads the authors to believe that there is little correlation between technical fibre stiffness and composite stiffness. A possible explanation for this could be that the composite behavior is dominated by the elementary fibres instead of the technical fibres. This can easily be understood from the fact that in a composite material, a matrix surrounds the the outer elementary fibres in the technical fibre. Consequently, the elementary fibres cannot deform freely in contrast to the single fibre test.

The strength of the composites seems to be well reflected by the technical fibre strength, although this should be verified by increasing the number of single fibre tests to reduce the variation on the technical fibre strength. Furthermore, fibre misalignment in the composites and the presence of impurities may “mask” this correlation, requiring detailed microscopic and tomographic inspection of the samples.

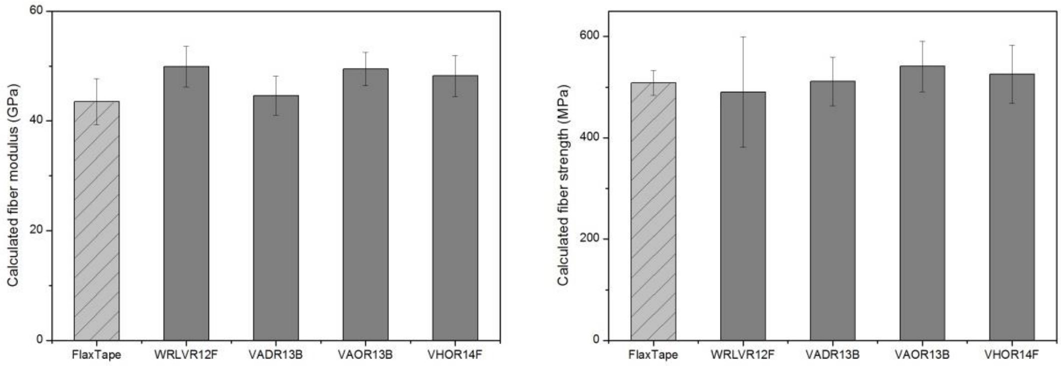


Figure 6: Left: Back-calculated fibre tensile modulus from the composite tensile tests. Right: Back-calculated fibre strength from the composite tensile tests.

3.3.2 Transverse flexion

The effect of fineness and purity on the transverse properties of the unidirectional composites was evaluated by three point bending tests. It is clearly seen from figures 7, 8 and 9 that the transverse flexural strength appears to be highly dependent on the fibre fineness and purity since all scutched fibre composites have a significantly lower transverse strength. In the hackled fibre composite, relatively more fibre surface is exposed to the matrix material than in the scutched fibres because of the difference in fineness. This automatically implies that fewer inter-elementary fibre interphases exist within the technical fibres. It is apparent that the hackling process eliminates the weakest of these interphases leading to fibres which are stronger in the transverse direction. Consequently, transverse failure is dominated by technical fibre failure which leads to the conclusion that the inter-elementary fibre interphase is weaker than the fibre-matrix interface in flax-epoxy composites.

Differences in transverse modulus were limited and only statistically different between the hackled material and the coarsest scutched fibres. This can be due to the presence of defects within the technical fibre bundles, thereby lowering the apparent transverse stiffness of the composite.

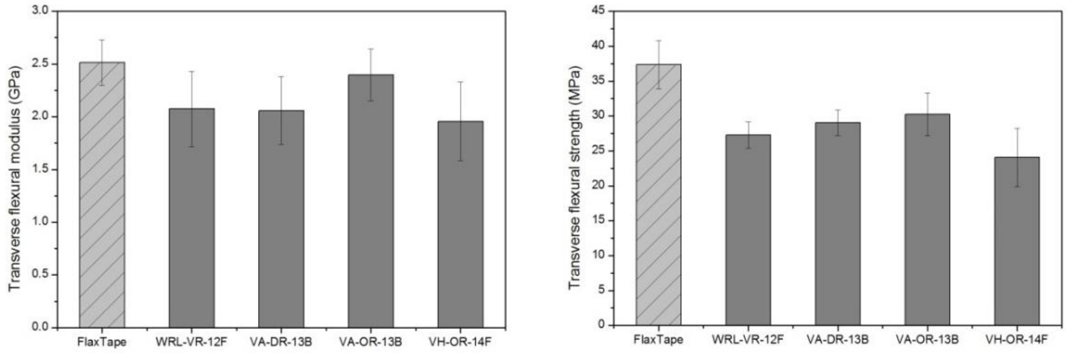
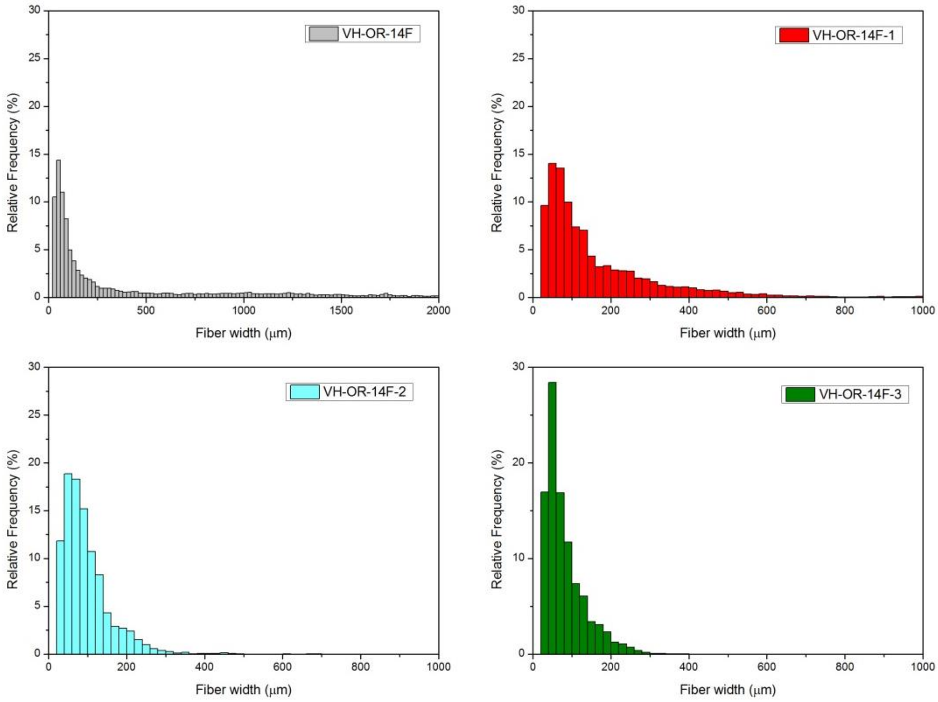


Figure 7: Left: Transverse flexural modulus of the composites. Right: Transverse flexural strength of the composites

To minimize compositional influences, the coarsest batch, OR1, was selected for further refinement by manual combing in 3 stages using a different comb in each stage, as described in section 2.3. The transverse flexural strengths of the composite samples, produced with fibres after each combing step, were determined.



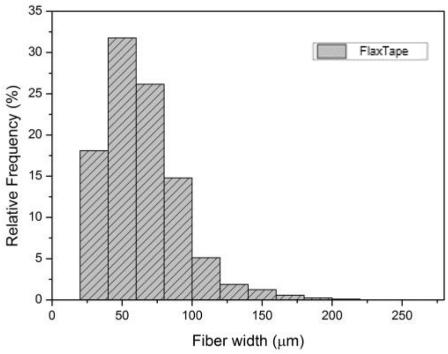


Figure 8: Fibre diameter distributions of the combed batch after each successive combing step. Scutched fibres are characterized by long tailed distributions. The fiber width distribution of the hackled material was added for comparison.

Figure 8 shows the results of the analysis on the OR1 batch after the first, second and third combing steps. The average diameters resulting from the distribution are mentioned in each histogram. It is clear that the as-received scutched fibres are characterized by a long tailed distribution leading to a high average fibre diameter. Combing, even with the coarsest comb, appears to significantly reduce the length of this tail. After the last combing step, the distribution approaches that of the hackled material.

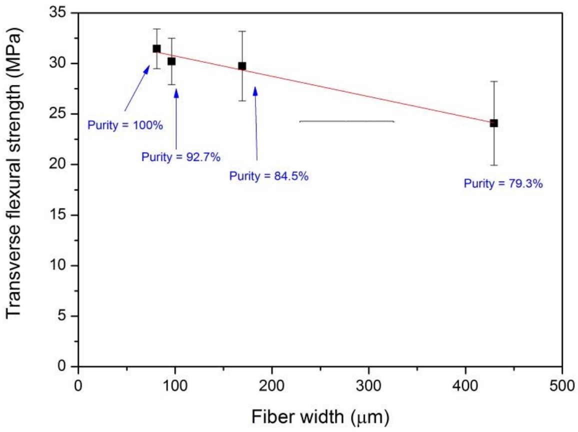


Figure 9: A strong negative correlation exists between transverse flexural strength and Ferret's diameter of the fibres.

Figure 9 shows that a negative correlation exists between Ferret's diameter determined by automated image analysis and transverse flexural strength within one batch. The correlation coefficient is -0.98. It appears that the change in transverse flexural strength is less sensitive to variations in purity since the correlation coefficient in this case was found to be only 0.85. However, these values should be interpreted with caution as they are the result of a limited amount of tests and the assumption that the third combing step yields a 100% pure material.

4 CONCLUSIONS

This work attempted to provide rapid screening methods for flax fibres in order to make it easier for flax fibre producers to judge the suitability of their flax for composite applications.

The determination of fibre fineness was investigated using gravimetric analysis and airflow, which are well established methods. Although the complexity of these techniques is low, sampling from the batches is difficult and may lead to significant errors. This may cause the results to be operator dependent. Secondly it was also found that the correlation between airflow and gravimetric results is low which lead to the suspicion that fineness is not the only factor influencing the end result in this technique.

Single fibre tensile tests carried out on technical flax fibres lead to the surprising result that scutched fibres behave in a non-linear way and tend to possess higher stiffness when compared to hackled fibres. This effect could be attributed to the increased amount of defects, such as kink bands, present in hackled fibres but this remains to be quantified.

Composite tensile tests showed little influence of the degree of fineness and purity on the longitudinal tensile performance of the composites. However, when the fibre stiffness was back-calculated from the stiffness of the composite, the values showed a large discrepancy with respect to what was obtained in technical fibre tests. This is an indication that technical fibre properties cannot be used to predict the performance of flax fibre composites.

Finally, fineness and purity have a large effect on the transverse composite properties, as shown in three point bending tests. The transverse flexural strength was found to be strongly correlated with fibre fineness. It was hypothesized that this correlation was due to the inherent weakness of the inter-elementary fibre interphase inside the technical fibres.

As a general conclusion, it is advised to only use composite testing to evaluate the suitability of flax fibres for composite applications. As was shown, transverse flexural strength of a unidirectional composite is indicative for the fibre fineness. Impregnated fibre bundle tests should always be carried out to determine if the tensile response of the flax fibres is sufficient for the envisioned application.

ACKNOWLEDGEMENTS

This work was performed in the scope of the FlaxPreComp project through the SIM Nanoforce program. The authors would like to thank all flax fibre producers in the consortium to provide the flax fibres for this work.

REFERENCES

[1] G. Coroller, A. Lefeuvre, A. Le Duigou, A. Bourmaud, G. Ausias, T. Gaudry, and C. Baley, "Effect of flax fibres individualisation on tensile failure of flax/epoxy unidirectional composite," *Compos. Part A Appl. Sci. Manuf.*, vol. 51, pp. 62–70, Aug. 2013.

[2] I. Van de Weyenberg, J. Ivens, a. De Coster, B. Kino, E. Baetens, and I. Verpoest, "Influence of processing and chemical treatment of flax fibres on their composites," *Compos. Sci. Technol.*, vol. 63, no. 9, pp. 1241–1246, 2003.

[3] I. Van de Weyenberg, T. Chi Truong, B. Vangrimde, and I. Verpoest, "Improving the properties of UD flax fibre reinforced composites by applying an alkaline fibre treatment," *Compos. Part A Appl. Sci. Manuf.*, vol. 37, no. 9, pp. 1368–1376, 2006.

[4] K. Charlet and A. Béakou, "Mechanical properties of interfaces within a flax bundle – Part I: Experimental analysis," *Int. J. Adhes. Adhes.*, vol. 31, no. 8, pp. 875–881, Dec. 2011.

[5] W. Hu, M. Zhang, M.-T. Ton-That, and T. Ngo, "A comparison of flax shive and extracted flax shive reinforced PP composites," *Fibers Polym.*, vol. 15, no. 8, pp. 1722–1728, 2014.

[6] H. L. Bos, "The potential of flax fibres as reinforcement for composite materials," Technische Universiteit Eindhoven, 2004.

[7] M. G. Callens, L. Gorbatikh, and I. Verpoest, "Tensile Behaviour of Ductile Steel Fibre / Epoxy Composites," in *DURACOSYS*, 2012, pp. 24–28.

[8] A. Lefeuvre, A. Bourmaud, C. Morvan, and C. Baley, "Elementary flax fibre tensile properties: Correlation between stress-strain behaviour and fibre composition," *Ind. Crops Prod.*, vol. 52, pp. 762–769, Jan. 2014.

[9] K. Charlet, J.-P. Jernot, M. Gomina, L. Bizet, and J. Breard, "Mechanical Properties of Flax Fibers and of the Derived Unidirectional Composites," *J. Compos. Mater.*, vol. 44, no. 24, pp. 2887–2896, Jul. 2010.

[10] C. Baley, "Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase," *Compos. Part A Appl. Sci. Manuf.*, vol. 33, no. 7, pp. 939–948, Jul. 2002.